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(54) **Separator plate for use in a gas fuel cell which comprises a set of electrodes, and also a stack of fuel cells**

Separatorplatte für Anwendung in einer Gas-Brennstoffzelle, enthaltend einen Satz von Elektroden, und auch eine Brennstoffzellenanlage

Plaque séparatrice pour utilisation dans une cellule à combustible gazeux comportant un ensemble d'électrodes, ainsi qu'une pile de cellules à combustible

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- **PATENT ABSTRACTS OF JAPAN, vol. 12, no. 273 (E-639)[3120], 29th July 1988; & JP-A-63 53 858**
- **PATENT ABSTRACTS OF JAPAN, vol. 12, no. 273 (E-639)[3120], 29th July 1988; & JP-A-63 53 857**
- **PATENT ABSTRACTS OF JAPAN, vol. 12, no. 130 (E-603)[2977], 21st April 1988; & JP-A-62 256 381**

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## Description

The invention relates to a separator plate for stacking fuel cells of the molten-carbonate type, provided with openings for fuel inlet and fuel outlet and for inlet and outlet of oxidizing agent, and also to a stack of fuel cells of the molten-carbonate type which contain said separator plates.

Such a separator plate is disclosed in the US Patent Specification 4,604,331 which describes a separator which is composed of a flat plate having two sealing flanges which are disposed on opposite sides and are bent back upwards and towards these themselves and are provided, at the two other sides, also situated opposite each other, of the plate with similar flanges which are, however, bent downwards. Each of the sealing flanges is composed of a flat wall which runs, at some distance from the plate, virtually parallel thereto and two side walls, folded in accordion fashion, of which one joins the flat wall to the plate and the other ends at a small distance from the plate, said side walls imparting a spring-type compressibility to the sealing flange in the direction generally perpendicular to the plane of the plate. Four corner elements close the ends of the sealing flanges.

US Patent Specification 4,514,475 discloses a separator which is composed of a flat rectangular gas-impermeable plate which is placed between two consecutive cells, two oppositely situated sides being folded back over one side of the plate to form two sealing flanges, the two remaining sides being folded back over the opposite side of the plate to form two other sealing flanges, each of the sealing flanges forming, together with the plate, a channel in which a spring-type compressible bundle of thin metal layers is placed. The two first sealing flanges adjoin the electrolyte matrix of one of the cells to form a gas-impermeable seal between an electrode of a cell and of a ribbed plate for reactant gas. The second sealing flange adjoins the electrolyte matrix of the other cell to form a gas-impermeable seal between electrodes of the other cell and of the other ribbed channels for the reactant gas. The sealing flanges interact with compressible stacks, joined thereto, of sheets for maintaining a distance between the plate and the electrolyte matrices, while they make it possible to adjust the distance between the plates.

An important characteristic of the separators according to these two patent specifications resides in the fact that, in both cases, the process gases for the different fuel cells of a stack have to be supplied externally via gas chambers, situated jointly opposite each other, for the fuel in the one direction and for the stream of oxidizing agent in a direction perpendicular thereto.

More generally, fuel cells are known for generating energy by electrochemical combustion of oxygen. The advantage of this mode of combustion is the direct conversion into electricity in addition to heat as a consequence of the internal resistance in the (fuel) cell. As a

result, process efficiencies of 60% and higher can be obtained. Although hydrogen is the fuel component in a fuel cell, it is also possible to use, inter alia, natural gas or coal gas as primary components.

The use of natural gas or coal gas requires conversion into a hydrogen-rich gas mixture by "reforming" or "chemical shift" reactions. These chemical reactions are endothermic and can be catalytically shifted at the working temperature of molten-carbonate fuel cell systems in the normal region of 600 to 700°C.

The molten-carbonate fuel cell is composed in principle of a plate-type anode, a matrix containing electrolyte and a cathode. This so-called active assembly of the fuel cell is enclosed on either side between one or more metal plates, termed current conductor and/or separator plate, which has to provide a number of functions such as: supporting the plate-type electrodes, passing the process gases over the electrodes, uniformly distributing the process gases over the electrode surface, conducting electrons between the separate fuel cells and separating the fuel gas and the oxidizing gas between anode and cathode of adjacent fuel cells. Figure 1 shows a basic diagram of a molten-carbonate fuel cell as described above.

In this figure, 1 indicates the matrix, for example an inert material of lithium aluminate, generally 0.5 to 1 mm thick, which is impregnated with a mixture of alkali carbonates. 2 is the cathode, for example nickel oxide, generally 0.4 to 0.8 mm thick and 3 represents the anode, for example nickel chromium, generally having a thickness of 0.6 to 0.8 mm. The brittle ceramic plate-type cathode is supported by a cathode current collector 4 which also distributes the oxidizing gas stream over the cathode and also conducts the electrons from the anode of the cell situated underneath via gas separation plate to the cathode 2.

The anode 3 is supported by the anode current collector 5 which also distributes the fuel cell gas stream over the anode and conducts the electrons from the anode via the separator plate to the cathode of the fuel cell situated above.

Hydrogen is supplied to the anode from the current collector situated at the anode side and  $H_2O + CO_2$  are removed therefrom.

$CO_3^{2-}$  is transported through the electrolyte matrix, while the reaction  $\frac{1}{2} O_2 + CO_2 + 2e$  giving  $CO_3^{2-}$  proceeds at the cathode. A mixture of  $O_2$  and  $CO_2$ , which may in turn originate partly from the anode side, is supplied to the cathode from the current collector situated at the cathode side. Figure 2 shows diagrammatically a possible practical embodiment of the cell construction of a molten-carbonate fuel cell described above having a half-cell adjoining at both sides. In this figure, 1 indicates the matrix, 2 is the cathode, 3 is the anode, 4 is the cathode current collector in the form of a corrugated plate, 5 is the anode current collector, 6 is the separator plate to the anode side of the fuel cell situated below, and 7 is the separator plate to the cathode side of the

fuel cell situated above. At 8 fuel is present and at 9 - oxidizing agent. A cell is therefore formed by the elements 1 to 5 inclusively, plus carbonate. The flow of the oxidizing gas and of the fuel gas over the cathode or over the anode respectively takes place perpendicularly to the plane of the drawing of Figure 2.

Above the separator plate 7, the adjacent half-cell situated above which has the cathode current collector 10, the cathode 11 and the matrix 12 is shown. In the same way, the adjacent half-cell situated below which has the anode current collector 13, the anode 14 and the matrix 15 is shown at the underside of separator plate 6.

In known cells, the interconnection of the cells is a problem. This is because, when the carbonate salt melts in the porous matrix and is received in the electrode, the volume "apparently" decreases. If the electrodes are interconnected by solid rails around the outside periphery, gaps are produced between the plate-type components as a result of shrinkage of the electrodes, as a result of which the performance of the cell declines and the efficiency is decreased or even, indeed, the operation of the cell stops completely.

A device of the type described in the preamble has now been found which is characterized in that the gas passages for the gas inlet and gas outlet are disposed in sides, situated opposite each other, of the separator plate. As a result, no sealing problems are produced by the carbonate melting and the receiving of the carbonate in the matrix and in the porous electrodes. In addition, as a result of the choice of a suitable profile of the gas passage, an elastic structure is obtained having the compression required to compensate for the shrinkage of the electrodes during the service life so that the internal resistance in the cell does not increase as a result of the formation of gaps and the performance of the cell is maintained.

A particular aspect of the discovery is the combination of the spring-loaded gas passage (or transfer region), described above, of the separator plate with a frame-type spring around the active cell assembly. Said "frame" spring provides the force required to seal off the process gases in the fuel cell from the environment. This frame spring may also contain built-in components for conveying the process gases.

Figure 3 shows, as an example, a possible embodiment of such a separator plate in which 16 shows the fuel inlet pipes and 17 the oxidizing agent inlet pipes, 18 and 19 show the corresponding outlet pipes, while 5 shows a corrugated plate-type current collector. The current collector plate is, of course, not bound by this embodiment.

Figure 4 shows a longitudinal section across the gas passage opening, in which B is the spring-loaded gas passage section and the supporting spring is situated around the active assembly in section C.

In this figure, 7 shows a partitioning plate or separator plate, 20 and 21 the anode gas container and cath-

ode gas container respectively, and 4 and 5 the current collectors, 2 being the cathode and 3 the anode, while the fuel inlet is 16 and the springs around the active cell assembly are indicated by 22. The function of these springs is to provide the required sealing force around the assembly in the central section of the cell. A suitable embodiment of the spring-loaded element around the active cell assembly has the form of a frame, a possible embodiment of which is shown in Figure 5, it being possible to achieve the mutual joints by, for example, a point weld or solder joint, said frame being provided with special devices for conveying the process flows. According to the invention, the spring action of the gas passage is achieved by a suitable shape of the outflow profile.

The spring characteristic of the spring beneath the active assembly is dimensioned in such a way that it is matched to the compression of the abovementioned gas passage and applies the required force to the matrix in order to achieve effective sealing of the process gases from the environment.

Figure 5 shows, at the points 25, a spot weld for fixing to the gas container (this may obviously also be positioned somewhat differently), 23 shows a bending line for a flow conveying device 26 and 24 shows the spring-loaded region. At the side edge, a possible embodiment of the spring construction at the anode side and the spring construction at the cathode side is shown.

Figures 6 and 7 show the unstressed height and the stressed height, that is to say built into the separator plate.

A stacking of fuel cells of said molten-carbonate type has now also been found which is characterized in that the cells are separated by said separator plates. The advantage is that such a stack of fuel cells is provided with a rail of spring construction around the active cell assembly.

Fuel cells of the molten carbonate type supply electricity at a relatively high current density of, for example, 150 to 160 A/cm<sup>2</sup>, but at a relatively low working voltage or cell voltage of approximately 0.7 - 0.8 V. For practical use, fuel cells of this type are used by stacking, for example, 100 fuel cells (fuel cell stacks). The function of the partitioning plates or separator plates under these circumstances is to separate the anodes and cathodes adjacent to each other, to separate the anode gas flow (fuel) and the cathode gas flow (oxidizing agent), to distribute the two gas flows uniformly over the electrodes, and to conduct electrons between the adjacent anode and cathode.

Figure 8 shows an assembly according to the invention in which 1 indicates the matrix, 2 the cathode, 3 the anode, 7 is a flat separator plate and 20 indicates a gas container, while 16 and 19 indicate, respectively, the process gas supplies, that is to say 16 indicates the fuel gas inlet, while 19 indicates the oxidizing agent outlet. The construction of the fuel cell stack is evident from this figure.

## Claims

1. Fuel cell of the molten carbonate type having separator plates (7) with a corrugated central portion (5) and gas-supplying and exiting openings on flat edges held between an upper and lower partitioning plate with corresponding openings, the partitioning plates (20,21) being provided with a transfer region between the flat partitioning plates and the separator plate, the connection between the transfer regions and the separator plate comprising a circumferential connection around said openings, characterized in that the transfer regions extend with an oblique angle in all directions both from the circumferential connection near the opening and the partitioning plate (20) to obtain a spring characteristic and in that spring means (22) are provided between the manifold plate and the separator plate (7), said spring means comprising a frame disposed around the active cell assembly, which frame-type spring contains built-in components for conveying the process gases, and wherein the profile of the frame spring is matched to the spring characteristic of the transfer region.
2. Stack of fuel cells of the molten carbonate type, characterized in that the cells are separated by separator plates as defined in claim 1.
3. Stack of fuel cells according to claim 2, characterized by a frame-type spring construction around the active cell assembly.

## Patentansprüche

1. Brennstoffzelle vom Typ aus geschmolzenem Karbonat, welche Trennplatten (7) aufweist, mit einem gewellten mittleren Bereich (5) und mit Gaszuführ- und Gasablaßöffnungen an flachen Seiten, welche zwischen einer oberen und einer unteren Unterteilungsplatte mit entsprechenden Öffnungen gehalten sind, wobei die Unterteilungsplatten (20, 21) mit einem Übertragungsbereich zwischen den flachen Unterteilungsplatten und der Trennplatte versehen sind, und wobei die Verbindung zwischen den Übertragungsbereichen und der Trennplatte eine Umfangsverbindung um die Öffnungen herum aufweist, **dadurch gekennzeichnet,** daß sich die Übertragungsbereiche in einem schiefen Winkel in alle Richtungen sowohl von der Umfangsverbindung nahe der Öffnung als auch von der Unterteilungsplatte (20) erstrecken, um eine Federkennlinie zu erhalten, und daß Federeinrichtungen (22) zwischen der Verteilerplatte und der Trennplatte (7) vorgesehen

sind, wobei die Federeinrichtungen einen Rahmen aufweisen, der um die Anordnung von aktiven Zellen herum angeordnet ist, wobei die rahmenartige Feder eingebaute Mittel zum Befördern der Prozeßgase enthält, und wobei der Querschnitt der Rahmenfeder der Federkennlinie des Übertragungsbereiches angepaßt ist.

2. Stapel von Brennstoffzellen vom Typ aus geschmolzenem Karbonat, **dadurch gekennzeichnet,** daß die Zellen durch die im Anspruch 1 definierten Trennplatten voneinander getrennt sind.
3. Stapel von Brennstoffzellen nach Anspruch 2, **gekennzeichnet durch** einen rahmenartigen Federaufbau, der um die Anordnung von aktiven Zellen herum angeordnet ist.

## Revendications

1. Cellule à combustible du type à carbonate fondu comportant des plaques séparatrices (7) ayant une partie centrale ondulée (5) et des ouvertures d'alimentation en gaz et de sortie des gaz formées sur des bords plats maintenus entre une plaque de répartition supérieure et une plaque de répartition inférieure ayant des ouvertures correspondantes, les plaques de répartition (20, 21) comportant une zone de transfert située entre les plaques de répartition plates et la plaque séparatrice, la liaison entre les zones de transfert et la plaque séparatrice comportant une liaison circonférentielle autour desdites ouvertures, caractérisée en ce que les zones de transfert s'étendent en formant un angle oblique dans toutes les directions à la fois à partir de la liaison circonférentielle à proximité de l'ouverture et de la plaque de répartition (20) pour obtenir une caractéristique élastique et en ce que des moyens élastiques (22) sont agencés entre la plaque de répartition et la plaque séparatrice (7), lesdits moyens élastiques comportant un cadre disposé autour de l'assemblage actif de cellules, lequel élément élastique du type cadre contient des composants intégrés pour transporter les gaz de service, et le profil du cadre élastique étant adapté à la caractéristique élastique de la zone de transfert.
2. Empilage de cellules à combustible du type à carbonate fondu, caractérisé en ce que les cellules sont séparées par des plaques séparatrices telles que définies dans la revendication 1.
3. Empilage de cellules à combustible selon la revendication 2, caractérisé en ce qu'il comporte une construction élastique du type cadre autour de l'assemblage actif de cellules.

fig - 1

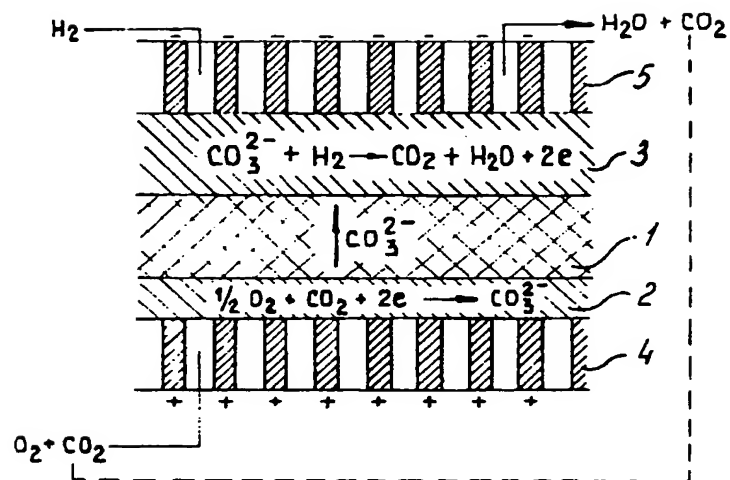


fig - 2

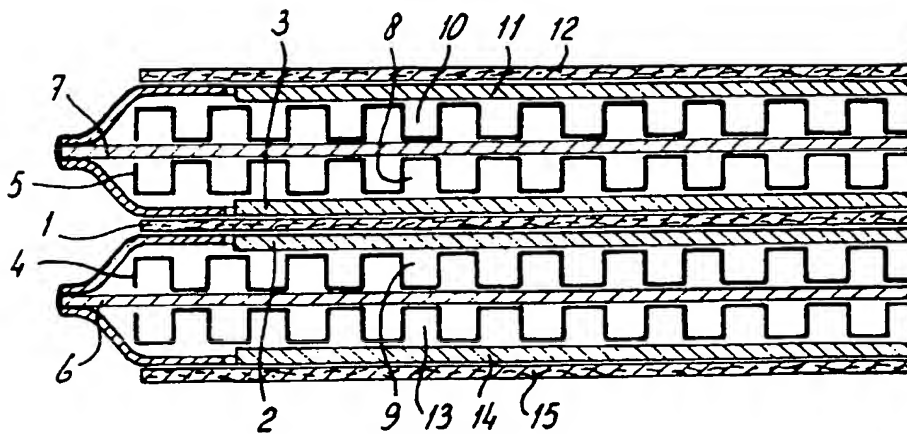


fig-3

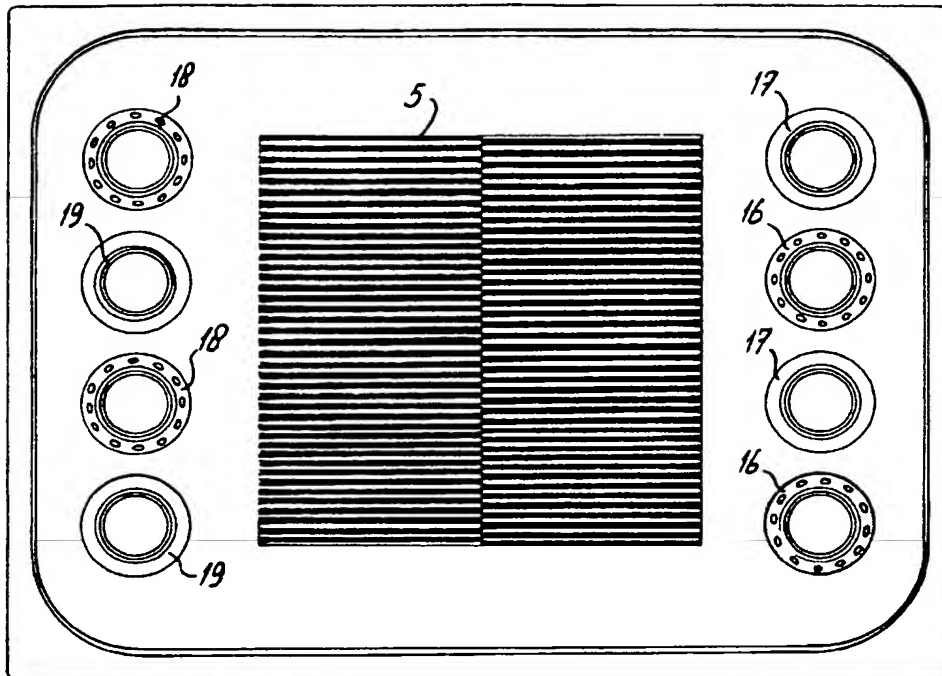


fig-4

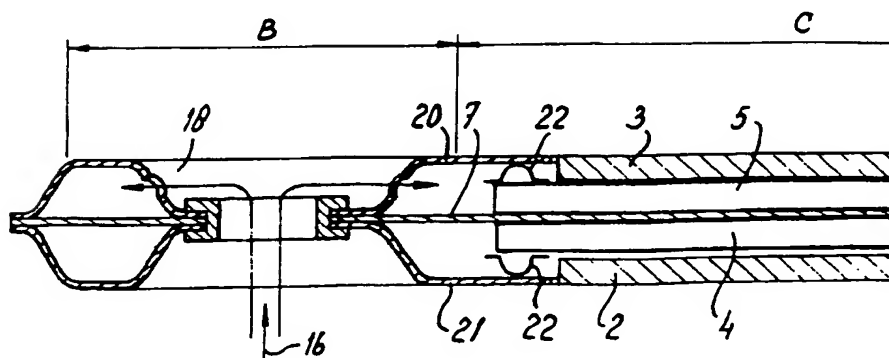


fig-5

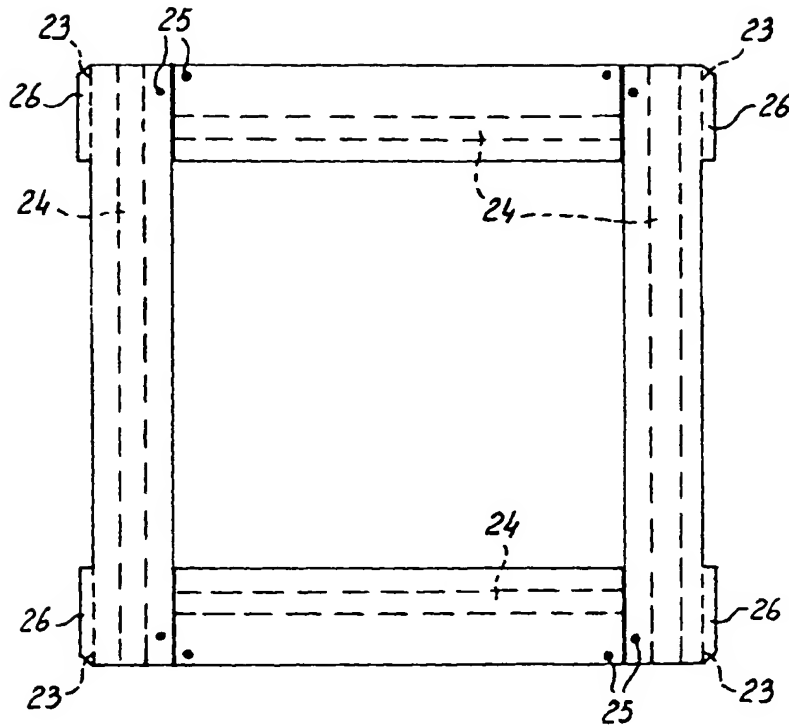


fig - 6

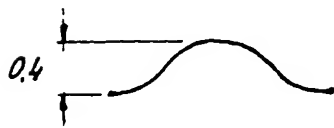


fig - 7

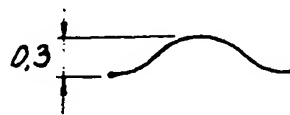


fig - 